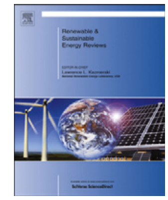
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Adsorber efficiency in adsorbtion refrigeration

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ABSTRACT

The coefficient of performance, COP, of an adsorption cooling is theoretically obtained. It shows the influence of the thermophysical properties of the adsorbent-adsorbate media regardless of the design of the machine. These quantities require a detailed understanding of the influence of temperature on the adsorbent and the adsorbate conditions in a cycle, so that an accurate determination of the thermodynamic behaviour is developed in order to obtain the COP.

Contents

1. Introduction

One of the main challenges of the industry today is to face its impact on global warming [1,2]; moreover, the greenhouse effect problem is not be solved completely yet. So, in addition to further developing the vapour compression technology, technical physicists and engineers have begun to explore new refrigeration technology such as solar cooling. Indeed, an adsorption cycle integrated into a solar energy system can be an alternative to a conventional system as a source of refrigeration [3]. This is generally more so in regions where solar flux is high and electric power is not reliably available to accommodate conventional cooling systems. Among the important applications of such refrigeration systems would be cooling for vaccine storage and for food storage [3,4]. On the contrary, in literature only a paper can be found on adsorption–desorption cycles on their thermodynamic theory [5].

The interest in the adsorption systems began in the 70s, during the oil crisis, for the new energy requirements, and continued in the 90s for the ecological problems related to the greenhouse gases. Today, the continue increase in energy consumption requires new efficient way of using energy resources [6]. Systems able to recover waste heat at low temperature can represent an interesting technology in relation to the environmental and energy use problems. Solid adsorbents (zeolites, activated carbon, silica gel, etc.) are porous materials and, at low temperatures, they can adsorb the refrigerant, while at high temperature they can release it: these materials can be used in refrigeration systems. The refrigeration cycle can be powered by thermal energy. Consequently, since the 80s, the basic principles and theory approach to solid adsorption refrigeration have been developed.

An adsorption refrigeration contains three components: container of adsorbents, condenser and evaporator. The adsorbent is packed in a hermetically sealed container. For solar adsorption refrigeration, for example, during the day, the high concentration of adsorbent and container are heated by solar radiation. At the maximum cycle temperature the refrigerant is desorbed from the adsorbent, its vapour condenses into liquid in the condenser, and heat is dissipated to the surroundings. From the condenser the liquid

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Nomenclature

Latin

A	area (m^2)
c	specific heat ($J\ kg^{-1}\ K^{-1}$)
COP	Coefficient of performance
F	free Helmholtz energy (J)
G	standard free energy (J)
H	enthalpy (J)
K	equilibrium constant
m	mass (kg)
n	moles (mol)
p	pressure (Pa)
Q	heat (J)
R	universal constant gas
T	temperature (K)
V	volume (m^3)

Greek

α	quantity which give the attraction between the molecules ($N\ m^{-1}$)
β	co-area of the adsorbed molecules (m^2)
γ	interface energy ($N\ m^{-1}$)
Γ	surface concentration ($mol\ m^{-2}$)
ϑ	covering degree
μ	chemical potential ($J\ mol^{-1}$)
ν	stechiometric coefficient
π	spreading pressure ($N\ m^{-1}$)

Superscript

0	isostearic
1	state 1 of the adsorption–desorption cycle
2	state 2 of the adsorption–desorption cycle
3	state 3 of the adsorption–desorption cycle
4	state 4 of the adsorption–desorption cycle
ads	adsorption
gas	gas
σ	interface
surf	surface

goes into the evaporator for gravity. During the night, the adsorbent is cooled to near ambient temperature: when the adsorbent pressure reaches the saturated vapour pressure of the refrigerant, the refrigerant boils in the evaporator and changes heat to the environment. The resulting refrigerant vapour is readsorbed into the adsorbent and cooling is produced [7]. The physical principle of operation is that the refrigerant concentration on the adsorbent depends on temperature under constant pressure conditions, so the adsorption and desorption occur by varying the temperature of their combination. So, a classical adsorption–desorption cycle is represented in Fig. 1 and it is composed by the following four processes [3]:

1. Heating and pressurisation ($1\to2$): the adsorber receives heat while being closed; the adsorbent temperature increases from T_1 to T_2, which induces a pressure increase, from the evaporation pressure p_1 up to the condensation pressure p_2.
2. Heating and desorption with condensation ($2\to3$): the adsorber continues receiving heat while being connected to the condenser, which now superimposes its pressure; the adsorbent temperature continues increasing form T_2 to T_3, which induces desorption of vapour; this desorbed vapour is liquefied in the condenser, while the pressure is constant, p_2.
3. Cooling and depressurisation ($3\to4$): the adsorber releases heat while being closed; the adsorbent temperature decreases from T_3 to T_4, which induces the pressure decrease from the condensation pressure down to the evaporation pressure to p_1.
4. Cooling and adsorption with evaporation ($4\to1$): the adsorber continues releasing heat while being connected to the evaporator, which now superimposes its pressure; the adsorbent temperature continues decreasing to T_1, which induces adsorption of vapour; this adsorbed vapour is vaporised in the evaporator; the evaporation heat is supplied by the heat source at low temperature.

If the thermal energy supply is the solar one, cycle is drive by the natural diurnal and nocturnal solar radiation time, so the adsorption cycles are intermittent because they are based on the heating–desorption–condensation phase and on the cooling–adsorption–evaporation phase but if the cooling system has two adsorbers, they can operate out of phase with the consequence of obtaining a quasi-continuous cold production.

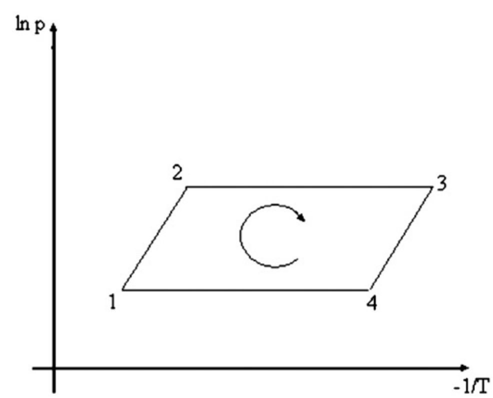


Fig. 1. A general adsorption–desorption cycle.

The aim of this paper is to discuss the coefficient of performance COP, useful to evaluate the cycle performance. Indeed, COP can show, for a given cycle, the influence of the thermophysical properties of the adsorbant–adsorbate media regardless of the design of the machine. These quantities in turn require a detailed and exact understanding of the influence of temperature on the adsorbant and the adsorbate conditions in a cycle. Consequently, in order to analyse the performance of an adsorption cycle, an accurate determination of the thermodynamic behaviour is needed under the expected operating conditions.

2. Kinds of adsorption cycles

The adsorption cooling systems may be classified in relation to the adsorbent and refrigerant combination used produced [7]

1. Adsorption cycles with water as the refrigerant: natural zeolite adsorbs large amounts of water vapour when cooled and desorbs the water vapour when heated [8]. This mineral, placed in a hermetically sealed metal container, performs the functions of energy collection and those of mechanical compressor in the conventional refrigerator [9]. The quantity of water adsorbed depends on temperature. Tchernev [9] first made a 100 dm^3

zeolite–water solar energy refrigerator with a collector/generator/absorber with 5 cm thick, 50 kg m^{-2} of zeolite: for a solar energy input of 6 kWh, the refrigerator produced 900 Wh/m^2 of cooling with a coefficient of performance (COP) of 0.15. The use of vapour/water is very interesting and promising in relation to the aim of use of the solar adsorption cooling.
2. Adsorption cycles with fluorocarbon as the refrigerant: Critoph [10] built a refrigerator to produce about 300 kJ of cooling. The solar collector/generator/absorber component comprised 9 lengths of 700 × 1 mm^2 wall thickness steel tube containing 24.2 kg of activated carbon, with a plan area of 1.02 m^2 coated with maxorb solar film. The refrigerator cooled from an ambient temperature of 30 °C down to 13 °C. This technology uses fluorocarbon refrigerants which are greenhouse gasses, so today it is not so interesting as the previous technology.
3. Adsorption cycles with ammonia as the refrigerant: always Critoph [11,12] made a carbon–ammonia refrigerator cycle. The generator, of exposed surface area 1.4 m^2, consisted of an array of 15 stainless steel tubes, each of 2 m length, 42 mm outside diameter and 1.1 mm thick, rated to 30 bar pressure, 17 kg of 208 °C activated carbon granules packed in the tubes, the condenser was 4 m in length and 12.5 mm diameter stainless steel tube coiled within a 100 l water tank, the evaporator was a 10 mm diameter stainless steel coil immersed in 4 l of water. The evaporator temperature attained was −1 °C and about 3 kg of ice was obtained, with a COP of 0.04. But this technologies requires high pressures and the ammonia is very corrosive.
4. Adsorption cycles with alcohol as the refrigerant: Pons and Guilleminot [13] used an activated carbon–methanol combination consisted of four solar collectors of total exposed area 6 m^2, containing 130 kg of adsorbent, two air cooled condensers and an evaporator with a net production of 30–35 kg of ice per sunny day, a COP in the range [0.10–0.12] for incident solar energy in the range [16,19] MJ/m^2. The activated carbon–methanol combination is preferred to other solid adsorbent–refrigerant combinations for its chemical stability, its enhanced COP, the inexpensive materials easily available in most developing countries the relative lightness of construction because the pressure inside is low.

In relation to the above summary, in solid adsorption solar refrigeration, three types of working materials are interesting

1. for adsorbate: ammonia, methanol and water;
2. for adsorbent: activated carbon, silica–gel and zeolite.

The adsorbate used must have some properties as follows [14]:

1. low cost;
2. easily available;
3. evaporation temperature below 0 °C;
4. small size of molecules such that it can be adsorbed into the adsorbent which must have micropores of diameter less than 20 Å;
5. high latent heat of vaporisation and low specific volume;
6. thermally stable with the adsorbent at the cycle operating temperature ranges;
7. non-toxic, non-corrosive and non-flammable;
8. low saturation pressures at normal operating temperature.

while the adsorbent must have the following:

1. adsorption of large amounts of the adsorbate under low temperature or desorption of most of the adsorbate when exposed to thermal energy;
2. high latent heat of adsorption compared to its sensible heating load;
3. no deterioration with age or use;
4. non-toxic and non-corrosive;
5. low cost;
6. easily available.

3. The chemical thermodynamic basis of adsorption

Adsorption is a process in which fluid molecules are trapped on the surface and within the pores

of a solid and it is a surface phenomenon [15]. The pore structure of adsorptive solids can be classified as

1. micropores for radii < 15 Å;
2. transitional pores for radii within 15–20 Å;
3. macropores for radii > 20 Å.

For refrigeration applications only the micropores are interesting. Two processes occur in solid adsorption [15]

1. The physical adsorption, characterised by low energy, is the result of the Van der Waals' forces of attraction and is predominant at low temperatures.
2. The chemisorptions, characterised by high energy, occur with chemical reaction.

Adsorption is the adhesion of atoms, ions, molecules of gas, liquid, or dissolved solids to a surface and during this process a film of the adsorbate on the surface of the adsorbent has origin. Adsorption is a consequence of surface energy. In a bulk material, all the bonding requirements of the constituent atoms of the material are filled by other atoms in the material. However, atoms on the surface of the adsorbent are not wholly surrounded by other adsorbent atoms and therefore can attract adsorbates. The exact nature of the bonding depends on the details of the species involved, but the adsorption process is generally classified as physical adsorption characteristic of weak Van der Waals forces, or chemisorption, characteristic of covalent bonding. It may also occur due to electrostatic attraction [15]. The chemical physical theoretical models of describing the adsorption do not completely describe this phenomenon in relation to the refrigeration because of the limited adsorbate concentration range required in cooling [14].

Now, in order to obtain a chemical thermodynamic approach to adsorption a global approach is developed, with the aim to use it in the evaluation of the COP of the solar powered adsorption cooling cycle.

The interface energy is defined as [15]

$$\gamma = \left(\frac{\partial F^s}{\partial A}\right)_{T,V^s,n_i^s} \tag{1}$$

where $F^s = \gamma A - pV^s + \sum_i \mu_i n_i^s$ is the interface free Helmholtz energy, A is the area of the surface, T is the temperature, V^s is the interface volume, n_i^s is the number of moles of the i-th component of the interface. The quantity γA is the free energy variation due to the formation of a surface dA, when temperature, volume and numbers of moles in the interface are constants. The adsorbed phase must satisfy the Gibbs–Duhem equation

$$dU = SdT - Vdp + \sum_i n_i d\mu_i \tag{2}$$

Consequently to the isothermal condition this last relation can be written as

$$Ad\gamma - V^s dp + \sum_i n_i^s d\mu_i = 0 \tag{3}$$

which, considering that the value of $V^s dp$ can be neglected in relation to the other ones, becomes

$$d\gamma = -\sum_i \Gamma_i d\mu_i \tag{4}$$

with $\Gamma_i = n_i^\sigma A^{-1}$. In the thermodynamic analysis of adsorption the spreading pressure π is defined as the pressure applied by the adsorbed substance on the solid surface [15]

$$p = \gamma_0 - \gamma = \frac{RT}{A-\beta} - \frac{\alpha}{A^2} \tag{5}$$

where γ_0 is the interface tension when there are no adsorption, R is the universal gas constant, A is the area of the adsorbent, β is the co-area of the adsorbed molecules and α is the quantity which give the attraction between the molecules. If we consider [15]

1. the gas is ideal then $dm_i = RT\ln p$
2. $dp = -d\gamma$

it follows that $(d\ln p/d\pi) = (1/\Gamma RT)$ and, from relation (5) we can obtain:

$$p = K\Gamma^0 \frac{\vartheta}{1-\vartheta} \exp\left(\frac{\vartheta}{1-\vartheta} - \frac{2\alpha\vartheta}{\beta RT}\right) \tag{6}$$

where $\vartheta = \Gamma/\Gamma^0$ is the covering degree of the surface as a function of the surface concentration Γ and the same quantity Γ^0 when a one layer is generated, $K = \exp(-\Delta G^0/RT) = K'\exp(-\Delta H^0/RT)$ is the equilibrium constant and $\Delta G^0 = \sum_i \nu_i \mu_i$ is the standard free energy variation, function only of temperature T, while ν_i is the stoichiometric coefficient of the i-th component of the chemical reaction, H^0 is the isosteric enthalpy in standard conditions whose behaviour in relation to ϑ is $\Delta H_{ads} = H^0 \ln\Gamma = H_{surf} - H_{gas}$ with H_{surf} the surface enthalpy and H_{gas} the gas enthalpy [15]. As a consequence the following relation can be obtained [15]:

$$\Delta H_{ads} = -RT^2 \left(\frac{\partial \ln p}{\partial T}\right)_\vartheta \tag{7}$$

From the relations (6) and (7) we can obtain the adsorption enthalpy variation

$$\Delta H_{ads} = -\Delta G^0 - \frac{2\alpha\vartheta}{\beta} - \frac{\Delta H^0}{R} \tag{8}$$

This quantity allows describing the adsorption phenomenon using a thermodynamic global approach. In the next section it will be used in the evaluation of the COP of the adsorber in adsorption refrigeration.

4. Adsorption refrigeration COP

The idea COP of the solar adsorption refrigeration is known in literature [14] as

$$COP = \frac{T_c(T_g - T_a)}{T_g(T_a - T_c)} \tag{9}$$

where T is the temperature and a means ambient, g generating and c condenser. Critoph developed the analysis of the COP and obtained another result [14]

$$COP = \frac{T_c}{T_g} \tag{9}$$

These COPs are related to ideal cycles and give information on the maximum value of the COP, but for real cycles it is necessary to obtain a theoretical expression useful to evaluate the COP. To do so, an adsorption–desorption cycle is considered as represented in Fig. 1. It has been described in the Section 1. The second principle analysis of this cycle can be developed as follows:

1. heating and pressurisation (1→2): the adsorber receives heat so that
$$Q_{12} = \Delta H_{12} = mc(T_2 - T_1) + V(p_2 - p_1) + \Delta H_{ads} \tag{10}$$
considering that the adsorption enthalpy variation ΔH_{ads} is negative as it will be discussed in the next section;
2. heating and desorption with condensation (2→3): the adsorber continues receiving heat so that
$$Q_{23} = \Delta H_{23} = mc(T_3 - T_2) \tag{11}$$
3. cooling and depressurisation (3→4): the adsorber releases heat so that
$$Q_{34} = \Delta H_{34} = mc(T_4 - T_3) + V(p_1 - p_2) - \Delta H_{ads} \tag{12}$$
4. cooling and adsorption with evaporation (4→1): the adsorber continues releasing heat
$$Q_{41} = \Delta H_{41} = mc(T_1 - T_4) \tag{13}$$

From this analysis it is possible to obtain

1. the heat obtained, Q_s
$$Q_s = Q_{12} + Q_{23} = \Delta H_{12} + \Delta H_{23} = mc(T_3 - T_1) + V(p_2 - p_1) + \Delta H_{ads} \tag{14}$$
2. the cooling heat Q_{ads}
$$Q_{ads} = Q_{34} = \Delta H_{34} = mc(T_4 - T_3) - V(p_2 - p_1) - \Delta H_{ads} \tag{15}$$

Consequently the COP is defined as [16–19]

$$\mathrm{COP} = \frac{Q_{ads}}{Q_s} \tag{16}$$

which becomes

$$\mathrm{COP} = \frac{mc(T_4 - T_3) - V(p_2 - p_1) - \Delta H_{ads}}{mc(T_3 - T_1) + V(p_2 - p_1) + \Delta H_{ads}} \tag{17}$$

which, considering the relation (8), becomes

$$\mathrm{COP} = \frac{mc(T_4 - T_3) - V(p_2 - p_1) + \Delta G^0 + \frac{2\alpha\vartheta}{\beta} + \frac{\Delta H^0}{R}}{mc(T_3 - T_1) + V(p_2 - p_1) - \Delta G^0 - \frac{2\alpha\vartheta}{\beta} - \frac{\Delta H^0}{R}} \tag{18}$$

which represents the theoretical COP for the adsorber in an adsorption cooling cycle.

5. Numerical evaluation

An innovative CO_2 based adsorption cooling system that amalgamates the behaviours of CO_2 adsorption and desorption on activated carbons is considered in relation to its detailed analysis developed by Jribi et al. [20]. The analysis presented is supported by the data reported by Jribi et al. [20]. They developed the analysis of the adsorption cooling process which used the adsorbent/adsorbate characteristics to obtain cooling at the evaporator by the combination of adsorption-triggered evaporation and desorption-resulted-condensation. The adsorption cooling system was composed by an evaporator, a reactor for thermal compression and a condenser; the pitch-based activated carbon (Maxsorb) and CO_2 were used as adsorbents and refrigerant. For continuous cooling operation, a low-pressure CO_2 refrigerant

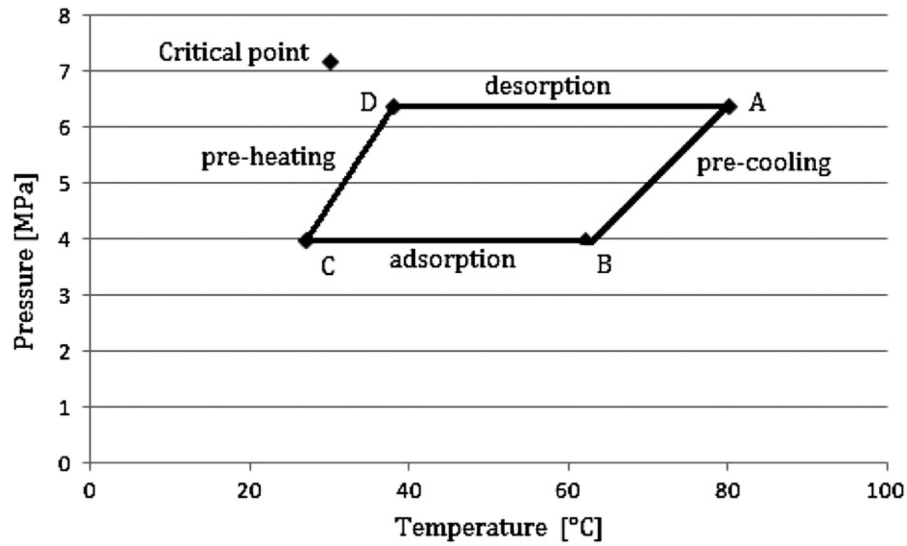


Fig. 2. Maxsorb-CO_2 adsorption cooling cycle [20].

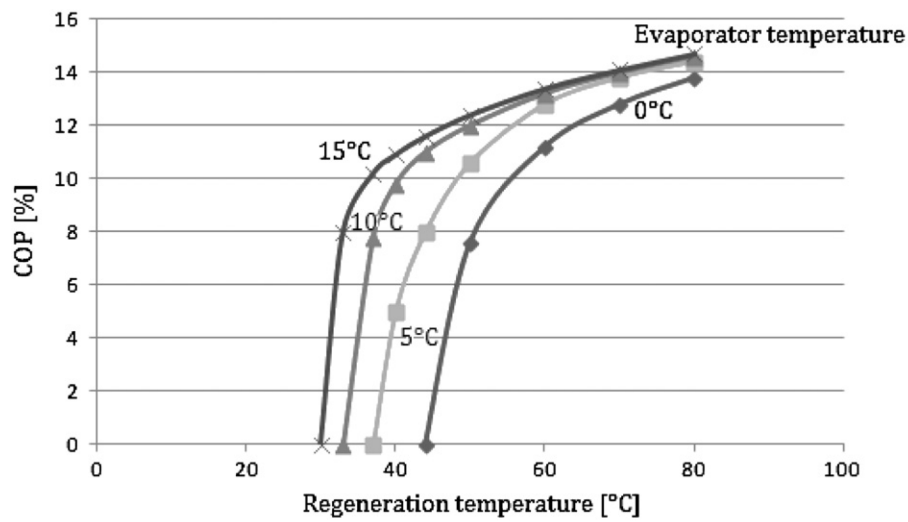


Fig. 3. Maxsorb-CO_2 adsorption cooling cycle COP.

was evaporated at the evaporator due to external cooling load and was adsorbed into the solid adsorbent in the adsorber at evaporator pressure (P_e). Then, the adsorbed bed was heated by an external heat source and the refrigerant was desorbed from the adsorbent and goes to the condenser at condenser pressure (P_c) for condensation by pumping heat through the environment. An average value of the isosteric heat of adsorption was taken 522.7 $kJkg^{-1}$ for the Maxsorb-CO_2 pair. Jribi et al. [20] summarised also the measurement of the adsorption equilibrium at three different temperatures 0 °C, 25 °C and 50 °C, and at pressures ranging from 0 to 5 MPa.

Using these data it has been possible to obtain a numerical evaluation of the relation (18). The cycle used by Jribi et al. [20] is summarised in Fig. 2, while the results here obtained for the COP are represented in Fig. 3. It is possible to state that the results are in accordance with the simulations of Jribi et al. [20].

6. Conclusions

In all European countries, there is a strong increase in the demand for building cooling and air-conditioning and is predicted still to increase in future. The reasons for this general increase are comfort habits, currently still low energy costs, architectural trends and slowly changing climate conditions. This rising demand for cooling and air-conditioning in buildings involves unfavourable fossil fuel consumptions as well as upcoming stability problems in the electricity supply in Mediterranean countries [21]. Indeed, for example the solar cooling can contribute to [21]:

1. replacement of fossil fuel demand by use of solar heat;
2. reduction of greenhouse effect emissions;
3. support in stability of electricity grids by less electricity energy and peak-power demand;
4. Optimised use of solar thermal systems through use of solar heat for combined assistance of space heating, cooling and domestic hot water preparation.

This paper has developed a discussion the coefficient of performance, useful to evaluate the cycle performance. The COP analysis has shown, for a given cycle, the influence of the thermophysical properties of the adsorbant-adsorbate media in relation to the operating conditions. The advantages in the use of this method can be summarised as follows:

1. it is a theoretical non ideal, but real, approach to evaluate the COP, and this can be useful in designing the cooling systems;
2. it considers the irreversibility in the evaluation of the chemical quantities introduced, based on the properties of the material used and also the specific cycle considered;
3. it agrees with the thermodynamic engineering approach found in literature, as shown in the previous numerical evaluation of this model.

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